

IN THE UNITED STATE PATENT AND TRADEMARK OFFICE

In re Patent application of

TAKENAKA et al.

Group Art Unit: 1625

5 Appln. No. 10/560,794

Examiner: Covington, Raymond K

Filed: December 15, 2005

Title: Curable Polycyclic Compounds and Process for the
production Thereof

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Declaration

I Hiromasa YAMAMOTO declare and state that:

1. I completed Department of Science, Graduate School of
15 Science, Osaka University [Master course] in March of 1987,
entered Tokuyama Soda K. K. (the name was changed to Tokuyama
Corporation in 1994) in April of 1987 as a researcher, was
assigned to Fujisawa Research Laboratory in May of 1987, was
assigned to Tsukuba Research Laboratories (the name was
20 changed to Advanced Materials Business Group in 2002) in
April of 1990, and has worked there until now.

In my assignments in Fujisawa Research Laboratory and
Tsukuba Research Laboratory, I worked mainly for the
development of organic materials (in particular, optical
25 materials and electronic materials) and am versed in the
technologies thereof.

2. I have reviewed and understand the contents of Dearborn
DN 48: 13253 (1953) abstract, Ciba DN 57: 75868 (1962)
30 abstract and the Office Action dated December 26, 2008 in the

subject application.

3. Purpose of Experiment

An adamantan compound in which an epoxy-containing group is introduced at the 1- and 3-positions of adamantan ring, can be produced by a method indicated in the present US Patent Application No. 10/560,794. The method is a method (Claim 19) of first reacting 1,3-adamantanediol with an alkali metal hydride to obtain an alcoholate, then reacting the alcoholate with an allyl halide to obtain 1,3-adamantanediol allyl ether, lastly oxidizing the allyl group to convert it to an epoxy group.

Meanwhile, Dearborn et al. (*Journal of Industrial and Engineering Chemistry*(1953),45,2715-21) disclose a method of reacting 1,5-dihydroxynaphthalene with epichlorohydrin in a presence of sodium hydroxide to produce 1,5-bis(2,3-epoxypropoxy)naphthalene.

However, the reactivity of the hydroxyl groups directly bonded to the 1- and 3-positions of 1,3-adamantanediol are extremely low as compared with the reactivity of the hydroxyl groups of 1,5-dihydroxynaphthalene.

Therefore, when 1,5-dihydroxynaphthalene is replaced by 1,3-adamantanediol according to the teaching of Dearborn et al., no reaction proceeds and it is impossible to produce an intended 1,3-bis(glycidoxoxy)adamantan compound.

The present experiment is intended to justify the above assertion.

4. Place where experiment was conducted

30 Tokuyama Corporation, Tsukuba Research Laboratories,

Experimental Room

40, Wadai, Tsukuba-shi, Ibaraki 300-4247, Japan

5. Period of Experiments

5 April 20 to May 12, 2009

6. Experiments

I have performed the following experiments.

Experiment 1

10 In a 100-ml, three-necked flask were placed 10 g (60 mmol) of 1,3-adamantanediol and 22.2 g (240 mmol) of epichlorohydrin. Thereto was dropwise added a 30% aqueous sodium hydroxide solution [1.6 g (0.04 mol) was dissolved in 3.7 g of water]. The mixture was stirred at 80°C for 4 hours.

15 During the reaction, the amount of 1,3-bis(glycidoxyoxy)adamantane formed was measured by gas chromatography. However, no intended 1,3-bis(glycidoxyoxy)adamantane was detected. After the completion of the reaction, there were recovered 8% of 1-glycidoxyoxy-3-adamantanol (a mono-substituted product) and 20 88% of 1,3-adamantanediol (raw material).

In order to enhance the reactivity, there was added, to the above reaction mixture, a 50% aqueous sodium hydroxide solution [1.6 g (0.04 mol) was dissolved in 1.6 g of water], followed by stirring at 90°C for 6 hours. The reaction was traced by gas chromatography; however, no intended 1,3-bis(glycidoxyoxy)adamantane was detected.

Experiment 2 The present Invention

30 A method of reacting 1,3-adamantanediol with an alkali metal

hydride to obtain an alcoholate and then reacting the alcoholate with an allyl halide.

5 Into a 200-ml, three-necked flask were fed 1.0 g (6.0 mmol) of 1,3-adamantanediol and 20 ml of dimethylformamide. Thereto was added 0.6 g (60 wt. %, 14.3 mmol) of sodium hydride, followed by stirring at 70°C for 2 hours. The reaction mixture was cooled to 5°C. Thereto was dropwise added 2.2 g (18.0 mmol) of allyl bromide. Violent foaming 10 occurred from the timing at which about the half amount was added dropwise. After the completion of the dropwise addition, stirring was conducted at 5°C for 5 hours. 50 ml of water was added and extraction was conducted using 100 ml of methylene chloride. The methylene chloride layer was 15 washed with 20 ml of water four times and the resulting organic layer was subjected to vacuum distillation to distil off the solvent. 1.7 g of a residue was obtained. Analysis of the residue indicated the presence of 94% of 1,3-bis(2-propenoxy)adamantane. The residue was purified by vacuum 20 distillation to obtain 1.2 g (4.8 mmol) of 1,3-bis(2-propenoxy)adamantane at a purity of 97%. Of the amount, 1.0 g (4.0 mmol) was fed into a 200-ml, three-necked flask and was dissolved in 50 ml of methylene chloride. Thereto was added 2.50 g (69% product, 10.0 mmol) of 25 m-chloroperbenzoic acid. Stirring was conducted at room temperature overnight. The resulting white precipitate was separated by filtration and the filtrate was washed with 10 ml of a saturated aqueous sodium sulfite solution. The absence of the oxidant in the organic layer was confirmed by 30 a potassium iodide-starch paper. The organic layer was

washed once with a 1 N aqueous sodium hydroxide solution and four times with water. The resulting organic layer was subjected to vacuum distillation to obtain 1.1 g of a residue. Analysis of the residue indicated the presence of 94% of 1,3-
5 bis(glycidyloxy)adamantane. The residue was purified by vacuum distillation to obtain 1,3-bis(glycidyloxy)adamantane of 97% purity in an amount of 0.9 g (3.2 mmol, yield: 53% (from 1,3-adamantanediol)).

10 I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and believe are believed to be true, and further that these statements are made with the knowledge that willful false statements and the like so made are punishable
15 by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

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Hiromasa Yamamoto
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Date: May 20, 2009